



(12) **EUROPEAN PATENT APPLICATION**

(43) Date of publication:  
04.06.1997 Bulletin 1997/23

(51) Int. Cl.<sup>6</sup>: **C10G 65/04**

(21) Application number: **96203362.7**

(22) Date of filing: **28.11.1996**

(84) Designated Contracting States:  
**DE FR GB IT NL**

(30) Priority: **28.11.1995 EP 95402713**

(71) Applicant: **SHELL INTERNATIONALE RESEARCH  
MAATSCHAPPIJ B.V.  
2596 HR Den Haag (NL)**

(72) Inventors:  
• **Bertaux, Jean-Marie Alexandre**  
**76530 Grand Couronne (FR)**  
• **Germaine, Gilbert Robert Bernard**  
**76530 Grand Couronne (FR)**  
• **Janssen, Martinus Maria Petrus**  
**76530 Grand Couronne (FR)**  
• **Hoek, Arend**  
**1031 CM Amsterdam (NL)**

(54) **Process for producing lubricating base oils**

(57) Process for the preparation of lubricating base oils having a VI of at least 150 from a Fischer-Tropsch wax feed, which process comprises the steps of:

- (a) contacting the Fischer-Tropsch wax feed with a hydroconversion catalyst under hydroconversion conditions,
- (b) separating the hydroconverted effluent obtained in step (a) into at least one lighter fraction and a heavy fraction, and
- (c) dewaxing the heavy fraction to yield the base oil,

wherein the Fischer-Tropsch wax feed has a congealing point of at least 50 °C and has such boiling range that the difference between the 90 %wt boiling point and the 10 %wt boiling point ( $T_{90}-T_{10}$ ) is in the range of from 40 to 150 °C.

10621829

## Description

The present invention relates to a process for producing lubricating base oils from Fischer-Tropsch waxes, and in particular lubricating base oils having a viscosity index (VI) of at least 150.

Processes for producing base oils having VI's above 150 from Fischer-Tropsch waxes are known in the art. For instance, in EP-A-0,515,256 a process for producing such base oils is disclosed, said process comprising the steps of:

- (a) contacting the synthetic or Fischer-Tropsch wax with hydrogen in the presence of an alumina-based hydroconversion catalyst;
- (b) contacting the effluent of step (a) with a hydro-isomerisation catalyst comprising a matrix, a specific zeolite Y and a hydrogenation component;
- (c) separating the effluent of step (b) into at least one lighter fraction and a heavy fraction; and
- (d) dewaxing the heavy fraction thereby yielding the base oil and a wax fraction. This wax fraction may be partly or totally recycled to hydro-isomerisation step (b).

The Fischer-Tropsch wax used as the feed in the working examples of EP-A-0,515,256 has a broad boiling range (difference between 90 %wt boiling point and 10 %wt boiling point is as high as 249 °C) which implies that a large variety of paraffinic molecules is present in said wax. Due to the presence of such large variety of different paraffinic molecules, it will be very difficult to meet the specifications as regards volatility when the base oils produced should be used as lubricating base oils.

In US-A-4,943,672 a process is disclosed for producing lubricating base oils having a VI of at least 130 from Fischer-Tropsch waxes, in which process the wax is first hydrotreated under severe conditions, then the hydrotreated wax is hydro-isomerised by contacting it with a fluorided Group VIII (noble) metal-on-alumina catalyst, subsequently the effluent from the hydro-isomerisation step is fractionated to produce a lubricating oil fraction and finally this lubricating oil fraction is dewaxed to produce the desired lubricating base oil. Unconverted wax recovered in the final dewaxing step may be recycled to the hydro-isomerisation step. The Fischer-Tropsch wax used should be a high boiling wax, since it is the intention to convert in the severe hydrotreating step that material present in the wax that has a boiling point above about 565 °C. In the Example of US-4,943,672 a Fischer-Tropsch wax is used which was obtained as the 370 °C+ fraction from the distillation of a Fischer-Tropsch synthesis product. Accordingly, the waxes used as the feedstocks are relatively high boiling waxes having a broad boiling range and a large heavy tail. Due to the broad boiling range and particularly due to the large heavy tail present in the wax, the fractionation after the hydroisomerisation step, or -if applied- after the

hydrofinishing step, should remove both the lightest fraction (boiling below 338 °C) and the heaviest fraction (boiling above 538 °C) from the hydroprocessed wax in order to obtain a final base oil product having acceptable volatility properties.

In US-A-5,059,299 a process for producing lubricating base oils having a VI of at least 130 and a pour point of -21 °C or lower from waxy feeds is disclosed, in which process the waxy feed -after an optional hydrotreating step- is first isomerised in an isomerisation zone at a predefined level of conversion, the total product of the isomerisation zone is then fractionated yielding a lube fraction boiling in the luboil range (i.e. above 330 °C and preferably above 370 °C) and this lube fraction is finally solvent dewaxed to yield the desired lubricating base oil and unconverted wax. This unconverted wax may be recycled to the isomerisation zone. The wax used as the feed may be a synthetic wax from a Fischer-Tropsch process or could be a slack wax obtained from a dewaxing process. No specific demands are made upon the Fischer-Tropsch waxes to be useful as a feed in the process disclosed. The Fischer-Tropsch wax used in Example 1 of US-A-5,059,299 is a high boiling wax having a relatively broad boiling range, which, as has already been stated above, results in the base oil product having unacceptable volatility properties. The isomerisation catalyst used suitably comprises a hydrogenating component on a halogenated refractory oxide support. The preferred catalyst is disclosed to be platinum on fluorided alumina.

Although the prior art processes perform satisfactory in many respects, there is still room for optimisation and improvement. The present invention aims to provide such improved process. More specifically, the present invention aims to provide a process for preparing base oils having a VI of at least 150 from a Fischer-Tropsch wax, which process involves a single hydro-processing stage and a fractionation stage, wherein only the lighter components need to be removed from the hydroprocessed effluent. Furthermore, it is an object of the present invention to provide base oils having excellent properties, particularly in terms of VI and volatility, at commercially attractive yields.

It has been found that these objects can be effectively achieved by using as the feed a specific Fischer-Tropsch wax having a relatively narrow boiling range and meeting certain requirements as to its congealing point.

Accordingly, the present invention relates to a process for the preparation of lubricating base oils having a VI of at least 150 from a Fischer-Tropsch wax feed, which process comprises the steps of:

- (a) contacting the Fischer-Tropsch wax feed with a hydroconversion catalyst under hydroconversion conditions,
- (b) separating the hydroconverted effluent obtained in step (a) into at least one lighter fraction and a heavy fraction, and

(c) dewaxing the heavy fraction to yield the base oil,

wherein the Fischer-Tropsch wax feed has a congealing point of at least 50 °C and has such boiling range that the difference between the 90 %wt boiling point and the 10 %wt boiling point ( $T_{90}-T_{10}$ ) is in the range of from 40 to 150 °C.

The Fischer-Tropsch wax used as the feed for the present process, is obtained via the well-known Fischer-Tropsch hydrocarbon synthesis process. In general, such Fischer-Tropsch hydrocarbon synthesis involves the preparation of hydrocarbons from a mixture of carbon monoxide and hydrogen at elevated temperature and pressure in the presence of a suitable catalyst. The Fischer-Tropsch catalyst normally is selective for preparing paraffinic molecules, mostly straight-chain paraffins, and the product from a Fischer-Tropsch synthesis reaction therefore usually is a mixture of a large variety of paraffinic molecules. Those hydrocarbons that are gaseous or liquid at room temperature are recovered separately, for instance as fuel gas ( $C_5^-$ ), solvent feedstocks and detergent feedstocks (up to  $C_{17}$ ). The more heavy paraffins ( $C_{18+}$ ) are recovered as one or more wax fractions, commonly referred to as Fischer-Tropsch wax(es) or synthetic wax(es). For the purpose of the present invention only those Fischer-Tropsch waxes are useful as the feed, which meet the aforementioned requirements with respect to their boiling range and congealing point.

Within the limits defined hereinbefore, preferred Fischer-Tropsch wax feeds are those having a congealing point in the range of from 55 to 150 °C, preferably from 60 to 120 °C and/or such boiling range that the  $T_{90}-T_{10}$  is in the range of from 50 to 130 °C. Those Fischer-Tropsch waxes melting below 100 °C, suitably have a kinematic viscosity at 100 °C ( $V_{k100}$ ) of at least 3 mm<sup>2</sup>/s, preferably between 3 and 12 mm<sup>2</sup>/s, more preferably between 4 and 10 mm<sup>2</sup>/s. Those Fischer-Tropsch waxes melting above 100 °C suitably have a kinematic viscosity at a temperature T, which is 10 to 20 °C higher than their melting point, in the range of from 8 to 15 mm<sup>2</sup>/s, preferably from 9 to 14 mm<sup>2</sup>/s.

The hydroconversion catalyst used in step (a) may in principle be any catalyst known in the art to be suitable for isomerising paraffinic molecules. In general, suitable hydroconversion catalysts are those comprising a hydrogenation component supported on a refractory oxide carrier, such as amorphous silica-alumina, alumina, fluorided alumina, molecular sieves (zeolites) or mixtures of two or more of these. One type of preferred catalysts to be applied in the hydroconversion step in accordance with the present invention are hydroconversion catalysts comprising platinum and/or palladium as the hydrogenation component. A very much preferred hydroconversion catalyst comprises platinum and palladium supported on an amorphous silica-alumina (ASA) carrier. The platinum and/or palladium is suitably present in an amount of from 0.1 to 5.0% by weight, more suitably from 0.2 to 2.0% by weight, calculated as

element and based on total weight of carrier. If both present, the weight ratio of platinum to palladium (calculated as element) may vary within wide limits, but suitably is in the range of from 0.05 to 10, more suitably 0.1 to 5. Examples of suitable noble metal on ASA catalysts are, for instance, disclosed in WO-A-94/10264 and EP-A-0,582,347. Other suitable noble metal-based catalysts, such as platinum on a fluorided alumina carrier, are disclosed in e.g. US-A-5,059,299 and WO-A-92/20759.

A second type of suitable hydroconversion catalysts are those comprising at least one Group VIB metal, preferably tungsten and/or molybdenum, and at least one non-noble Group VIII metal, preferably nickel and/or cobalt, as the hydrogenation component. Usually both metals are present as oxides, sulphides or a combination thereof. The Group VIB metal is suitably present in an amount of from 1 to 35% by weight, more suitably from 5 to 30% by weight, calculated as element and based on total weight of catalyst. The non-noble Group VIII metal is suitably present in an amount of from 1 to 25 %wt, preferably 2 to 15 %wt, calculated as element and based on total weight of carrier. A hydroconversion catalyst of this type which has been found particularly suitable is a catalyst comprising nickel and tungsten supported on fluorided alumina.

A third class of suitable hydroconversion catalysts are those based on an intermediate pore size zeolitic material, suitably comprising at least one Group VIII metal component, preferably Pt and/or Pd, as the hydrogenation component. Suitable zeolitic materials, then, include ZSM-5, ZSM-22, ZSM-23, ZSM-35, SSZ-32, ferrierite, zeolite beta, mordenite and silica-alumino-phosphates, such as SAPO-11 and SAPO-31. Examples of suitable hydroisomerisation catalysts are, for instance, described in WO-A-92/01657.

The hydroconversion conditions applied in step (a) are those known to be suitable in hydro-isomerisation operations. Suitable conditions, then, involve operating temperatures in the range of from 275 to 450 °C, preferably 300 to 425 °C, a hydrogen partial pressure in the range of from 10 to 250 bar, suitably 25 to 200 bar, a weight hourly space velocity (WHSV) in the range of from 0.1 to 10 kg/l/h, preferably 0.2 to 5 kg/l/h, and a gas rate in the range of from 100 to 5,000 NI/kg, preferably 500 to 3,000 NI/kg.

In step (b) of the present process the hydroconverted effluent from step (a) is separated into at least one lighter fraction and a heavy fraction. The effective cutpoint of the heavy fraction is suitably in the range of from 325 to 450 °C and even more suitably is in the range of from 350 to 420 °C, particularly when the lubricating base oils to be obtained are to be used in engine oils. The effective cutpoint of the heavy fraction is the temperature above which at least at least 85% by weight and preferably at least 90% by weight, of the hydrocarbons present in this heavy fraction has its boiling point. This separation or fractionation can be achieved by techniques known in the art, such as

atmospheric and vacuum distillation or vacuum flashing.

The heavy fraction obtained in step (b) is subsequently subjected to a dewaxing treatment in step (c) to arrive at the desired pour point. The dewaxing carried out in step (c) may in principle be carried out via any known dewaxing process. Examples of suitable dewaxing operations are the conventional solvent dewaxing processes, particularly those wherein methylethylketone, toluene or a mixture thereof is used as the dewaxing solvent, and the catalytic dewaxing processes. Both types of dewaxing operations are well known in the art. The most commonly applied solvent dewaxing process is the methyl ethyl ketone (MEK) solvent dewaxing route, wherein MEK is used as the dewaxing solvent, possibly in admixture with toluene. Catalytic dewaxing generally involves cracking and/or isomerising linear and slightly branched paraffinic hydrocarbon molecules -which negatively influence the cold flow properties of the base oil- in the presence of hydrogen and a dewaxing catalyst under appropriate dewaxing conditions. Suitable dewaxing catalysts which mainly favour cracking of paraffinic hydrocarbons are those comprising ZSM-5, ferrierite and/or silicalite and optionally a hydrogenation component. Examples of catalysts which mainly favour isomerisation of linear or slightly branched hydrocarbons, include catalysts comprising a silicoaluminophosphate (SAPO), such as e.g. SAPO-11, SAPO-31 and SAPO-41, ZSM-23 and SSZ-32. Another class of suitable dewaxing catalysts for use in dewaxing step (c) are those catalysts based on a molecular sieve having pores with a diameter in the range of from 0.35 to 0.80 nm and containing covalently bound alumina moieties in its framework, which molecular sieve has been modified to reduce the mole percentage of alumina, suitably by a surface dealumination treatment. This type of catalysts and dewaxing operations wherein such catalysts are used, are disclosed in European patent application No. 95401379.3/ref. TS 5518). Accordingly, a particularly suitable class of dewaxing catalysts comprise a hydrogenation component supported on surface deactivated molecular sieve and optionally a low acidity refractory oxide binder material. The hydrogenation component may comprise at least one Group VIB metal component (e.g. one or more of tungsten, molybdenum and chromium) and/or at least one Group VIII metal component (e.g. one or more of palladium, platinum, nickel and cobalt). It has been found particularly preferred for the purpose of the present invention to employ a hydrogenation component comprising platinum and/or palladium, suitably present in an amount of from 0.2 to 3.0% by weight (calculated as element and based on total weight of support, i.e. modified molecular sieve plus optional binder). Suitable molecular sieves include MFI-type zeolites, such as ZSM-5 and silicalite, offretite, ferrierite, ZSM-35 and zeolites of the MTT-type, such as ZSM-23 and SSZ-32. Of these, the MTT-type zeolites, ferrierite, ZSM-5 and mixtures thereof are preferred for the purpose of

the present invention. If present at all, suitable binder materials include low acidity refractory oxides such as silica, zirconia, titanium dioxide, germanium dioxide, boria and mixtures of two or more of these with silica being most preferred. The weight ratio of surface deactivated molecular sieve to binder may range from 10/90 to 100/0.

The slack wax obtained in the dewaxing treatment of step (c) is suitably recycled, i.e. all or part of this slack wax is routed back to the hydroconversion step (a), most conveniently by blending it with the fresh Fischer-Tropsch wax feed. In this way the final yield of lubricating base oil can be maximised.

The lubricating base oils obtained by the process according to the present invention can be used in a variety of oils. For instance, those lubricating base oils obtained from Fischer-Tropsch waxes having a  $T_{90}$  between about 400 and 500 °C are very useful in electrical oils, transformer oils and refrigerator oils. Those base oils obtained from Fischer-Tropsch waxes having a  $T_{90}$  above 450 °C, suitably between 450 and 575 °C, are very useful as lubricating base oils used for the more sophisticated lubricants required in, for instance, automotive engines.

The invention is now further illustrated by the following examples without restricting the scope of the invention to these specific embodiments.

#### Example 1

A Fischer-Tropsch wax having the properties as listed in Table I was contacted with a fluorided NiW/alumina catalyst (5.0 %wt Ni, 23.1 %wt W, 4.6 %wt F, all based on total weight of carrier) at a temperature of 383 °C, a hydrogen partial pressure of 140 bar, a WHSV of 1 kg/l/h and a gas rate of 1,500 NI/kg. The effluent was fractionated and the 390 °C+ fraction (obtained at a yield of 87.8% by weight based on total effluent) was subsequently solvent dewaxed using MEK/toluene at -20 °C. The resulting base oil had a VI of 165, a pour point of -15 °C, a kinematic viscosity at 100 °C (Vk100) of 4.95 mm<sup>2</sup>/s and a Noack volatility (as determined by CEC-L-40-T87) of 8.3% by weight. Total yield of lubricating base oil amounted up to 41% by weight based on Fischer-Tropsch wax feed.

TABLE I

| Properties of Fischer-Tropsch wax     |      |
|---------------------------------------|------|
| T <sub>10</sub> (°C)                  | 432  |
| T <sub>50</sub> (°C)                  | 482  |
| T <sub>90</sub> (°C)                  | 527  |
| T <sub>90</sub> -T <sub>10</sub> (°C) | 95   |
| CPI (°C)                              | 69   |
| Vk100 (mm <sup>2</sup> /s)            | 5.67 |

**Example 2**

The same Fischer-Tropsch wax as used in Example 1 was contacted with a PtPd/ASA (0.3 %wt Pt, 1 %wt Pd, ASA: silica/alumina molar ratio is 55/45) catalyst at a temperature of 332 °C, whilst the other conditions were the same as applied in Example 1. The effluent was fractionated and the 390 °C+ fraction (obtained at a yield of 88.3% by weight based on total effluent) was subsequently solvent dewaxed using MEK/toluene at -20 °C. The resulting base oil had a VI of 167, a pour point of -15 °C, a kinematic viscosity at 100 °C (Vk100) of 4.86 mm<sup>2</sup>/s and a Noack volatility of 7.4% by weight. Total yield of lubricating base oil amounted up to 39% by weight based on Fischer-Tropsch wax feed.

**Example 3**

The procedure of Example 2 was repeated except that the 390 °C+ fraction obtained was catalytically dewaxed instead of solvent dewaxed. Catalytic dewaxing was carried out by passing said 390 °C+ fraction over a dewaxing catalyst comprising 0.7% by weight of Pt on silica-bound surface dealuminated ZSM-23 (70 %wt surface dealuminated ZSM-23, 30 %wt silica; surface dealumination carried out according to the method disclosed in U.S. Patent No. 5,157,191 using ammonium hexafluorosilicate) at a temperature of 310 °C, a hydrogen partial pressure of 40 bar, a WHSV of 1 kg/l/h and a gas rate of 693 NI/kg.

The resulting base oil had a VI of 151, a pour point of -27 °C, a kinematic viscosity at 100 °C (Vk100) of 4.96 mm<sup>2</sup>/s and a Noack volatility (as determined by CEC-L-40-T87) of 8.8% by weight. Total yield of lubricating base oil amounted up to 62.4% by weight based on Fischer-Tropsch wax feed.

**Claims**

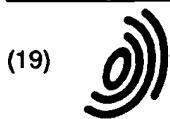
1. Process for the preparation of lubricating base oils having a VI of at least 150 from a Fischer-Tropsch

wax feed, which process comprises the steps of:

- (a) contacting the Fischer-Tropsch wax feed with a hydroconversion catalyst under hydroconversion conditions,
- (b) separating the hydroconverted effluent obtained in step (a) into at least one lighter fraction and a heavy fraction, and
- (c) dewaxing the heavy fraction to yield the base oil,

wherein the Fischer-Tropsch wax feed has a congealing point of at least 50 °C and has such boiling range that the difference between the 90 %wt boiling point and the 10 %wt boiling point (T<sub>90</sub>-T<sub>10</sub>) is in the range of from 40 to 150 °C.

2. Process according to claim 1, wherein T<sub>90</sub>-T<sub>10</sub> is in the range of from 50 to 130 °C.
3. Process according to claim 1 or 2, wherein the Fischer-Tropsch wax feed has a congealing point in the range of from 55 to 150 °C, preferably from 60 to 120 °C.
4. Process according to any one of the preceding claims, wherein the hydroconversion catalyst comprises a hydrogenation component supported on a refractory oxide carrier.
5. Process according to claim 4, wherein the hydroconversion catalyst comprises platinum and/or palladium as the hydrogenation component.
6. Process according to claim 5, wherein the hydroconversion catalyst comprises platinum and palladium supported on an amorphous silica-alumina carrier.
7. Process according to claim 4, wherein the hydroconversion catalyst comprises at least one Group VIB metal, preferably tungsten, and at least one non-noble Group VIII metal, preferably nickel, as the hydrogenation component.
8. Process according to any one of the preceding claims, wherein the heavy fraction is obtained in step (b) at an effective cutpoint in the range of from 350 to 420 °C.
9. Process according to any one of the preceding claims, wherein the dewaxing in step (c) is carried out by solvent dewaxing.
10. Process according to any one of claims 1 to 8, wherein the dewaxing in step (c) is carried out by catalytic dewaxing.



Europäisches Patentamt

European Patent Office

Office européen des brevets



(11) **EP 0 776 959 A3**

(12) **EUROPEAN PATENT APPLICATION**

(88) Date of publication A3:  
11.03.1998 Bulletin 1998/11

(51) Int. Cl.<sup>6</sup>: **C10G 65/04**

(43) Date of publication A2:  
04.06.1997 Bulletin 1997/23

(21) Application number: 96203362.7

(22) Date of filing: 28.11.1996

(84) Designated Contracting States:  
**DE FR GB IT NL**

(30) Priority: 28.11.1995 EP 95402713

(71) Applicant:  
**SHELL INTERNATIONALE RESEARCH  
MAATSCHAPPIJ B.V.  
2596 HR Den Haag (NL)**

(72) Inventors:

- Bertaux, Jean-Marie Alexandre  
76530 Grand Couronne (FR)
- Germaine, Gilbert Robert Bernard  
76530 Grand Couronne (FR)
- Janssen, Martinus Maria Petrus  
76530 Grand Couronne (FR)
- Hoek, Arend  
1031 CM Amsterdam (NL)

(54) **Process for producing lubricating base oils**

(57) Process for the preparation of lubricating base oils having a VI of at least 150 from a Fischer-Tropsch wax feed, which process comprises the steps of:

- (a) contacting the Fischer-Tropsch wax feed with a hydroconversion catalyst under hydroconversion conditions,
- (b) separating the hydroconverted effluent obtained in step (a) into at least one lighter fraction and a heavy fraction, and
- (c) dewaxing the heavy fraction to yield the base oil,

wherein the Fischer-Tropsch wax feed has a congealing point of at least 50 °C and has such boiling range that the difference between the 90 %wt boiling point and the 10 %wt boiling point ( $T_{90}-T_{10}$ ) is in the range of from 40 to 150 °C.

EP 0 776 959 A3



European Patent  
Office

# EUROPEAN SEARCH REPORT

Application Number  
EP 96 20 3362

| DOCUMENTS CONSIDERED TO BE RELEVANT   |   |   |  |
|---|---|---|--|
| Category  | Citation of document with indication, where appropriate, of relevant passages | Relevant to claim                                   | CLASSIFICATION OF THE APPLICATION (Int.Cl.6) |
| D,A   | EP 0 515 256 A (INST FRANCAIS DU PETROL)<br>* the whole document *            | 1-10  | C10G65/04                                    |
| D,A   | US 4 943 672 A (HAMNER DECEASED GLEN P ET AL)<br>* the whole document *       | 1-10  |  |
| D,A   | US 5 059 299 A (CODY IAN A ET AL)<br>* the whole document *                   | 1-10  |  |
| A   | EP 0 668 342 A (SHELL INT RESEARCH)<br>* the whole document *                 | 1-10  |  |
| A   | EP 0 464 547 A (MOBIL OIL CORP)<br>* the whole document *                     | 1-10  |  |
| A   | EP 0 574 191 A (MOBIL OIL CORP)<br>* the whole document *                     | 1-10  |  |
| A   | EP 0 471 524 A (EXXON RESEARCH ENGINEERING CO)<br>* the whole document *      | 1-10  | TECHNICAL FIELDS SEARCHED (Int.Cl.6)         |
|   |   |   | C10G   |
| The present search report has been drawn up for all claims  |   |   |  |
| Place of search<br>THE HAGUE  |   | Date of completion of the search<br>19 January 1998 | Examiner<br>Michiels, P                      |
| <p>CATEGORY OF CITED DOCUMENTS</p> <p>X : particularly relevant if taken alone<br/>Y : particularly relevant if combined with another document of the same category<br/>A : technological background<br/>O : non-written disclosure<br/>P : intermediate document</p> <p>T : theory or principle underlying the invention<br/>E : earlier patent document, but published on, or after the filing date<br/>D : document cited in the application<br/>L : document cited for other reasons<br/>&amp; : member of the same patent family, corresponding document</p> |   |   |  |

EPO FORM 1503 03.82 (P4/C01)

